MEASUREMENT OF RADON GAS CONCENTRATION IN FERTILIZER SAMPLES USING IRRADIATED CR-39 NUCLEAR TRACK DETECTOR

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ABSTRACT

In the present work, we have measured the radon gas concentration; effective dose and exhalation rate for some fertilizers samples, raw materials, triple super phosphate and single super phosphate by using alpha-emitters registrations that are emitted from radon gas in CR-39 nuclear track detector after irradiated with gamma rays. The results indicated that the highest average radon gas concentration in fertilizer samples was found in raw material, triple super phosphate (T.S.P) samples (269.620Bq/m^3 and 186.8 Bq/m^3, respectively) while the lowest average radon gas concentration was found in single super phosphate (S.S.P.) for powder and granules samples, 79.6 Bq/m^3 and 52.90 Bq/ m^3, respectively . These results show that the radon concentration; effective dose and exhalation rate in all fertilizers samples and waste product are below the allowed limit according to the International Commission of Radiation Protection agency.

Key words: Gamma irradiation, Alpha particle, CR-39.

INTRODUCTION

Radiation processing is a useful technology to induce suitable modifications of materials. The interest in radiation treatment of polymers has increased, prompted by the radiations induced modifications of the properties of various polymeric materials. The physical and chemical modifications induced in polymers under radiation are triggered by the energy loss by the radiation within the target. Solid state nuclear track detectors (SSNTD) are used widely in several technical applications for the detection of charged particles from protons to heavy ions, as well as the simple registration of the particle flux density or the fluencies in the environmental dosimetry (Hermsdorf *et al*., 2007). On the other hand, phosphate ore is the raw material used in production of some fertilizers. It contains various amounts of natural radioactive elements. During the phosphate ore processing, owing to chemical properties of Radium, practically all ^{226}Ra gets incorporated into phosphorgypsum and remains in disequilibrium status when compared to radioactivity levels contained in the raw material. Most of the phosphogypsum is considered waste and is stockpiled or discharged into the aquatic environment (UNSCAR 2000). Potential issues of concern resulting from phospho-gypsum disposal are its environmental impacts; possible increases in radio- nuclides in soils or in groundwater and consequential ingestion by humans through exposure routes such as drinking water and food chain (Laich, 1995). Once, deposited in bone tissue; ^{226}Ra has a high potential for causing biological damage through continuous irradiation of human skeleton over many years and may induce bone sarcoma (Marovic *et.al*., 1995). The natural radionuclides of concern are mainly Potassium, Uranium, Thorium, and the radio-nuclides that are created as their radioactive decay chains. Emanation of Radon ^{222}Rn and ^{220} Rn of half life time's 3.8 d and 65 s into air occurs as a product of uranium ^{238}U and thorium ^{232}Th decay chains, respectively. The short lived decay products of radon are responsible for most of the hazards by inhalation. The hazard of Radon comes from its radioactive progeny, which use their physical properties to spread or attach like aerosols, trapped in the lung and depositing their alpha-particle energies in the tissue, producing higher ionization density than beta particles or gamma-rays. Lung & skin cancer, and kidney diseases are the health effects attributed to inhalation of radon-decay products (Kumar, 1986). The sources of radon gas are the building materials and its components, ground water, and soil (*Ahmad, et.al, 1998)*. The radiological impact from the above nuclides is due to radiation exposure of the body by the gamma rays and irradiation of the lung tissues from inhalation of Radon and its progeny (Papastefanou, *et.al*; 1983). From the natural risk point of view, it is necessary to know the dose limits of public exposures and to measure the natural environmental radiation level provided by ground, air, water, foods, building interiors etc., for the estimation of the exposures to natural radiation sources. (IAEA; 1989). The aim of the present study is not only to enhance the dosimetric properties of CR-39 but also to use it in the determination of radon concentration level in some fertilizer samples and some of its products besides the annual effective dose (AED).

MATERIAL AND METHODOLOGY

Material:

The present investigation is based on the study of different kinds of fertilizer samples which was available in the local market, some of them are products from factory A in Abo Zabal area and another factory B present in Kafer Al Zyat area. The samples under investigation were in four forms, raw material, triple super phosphate (T.S.P), single super phosphate (S.S.P), beside waste products. The determination of the concentrations of alpha particles emitted from radon gas in fertilizer samples were performed by using enhanced CR-39 nuclear track detector of thickness (500 μm) which was irradiated with 20 Gy.

Methodology:

Pre gamma irradiation:

Samples from CR-39 sheets were irradiated using CO^{60} gamma source in the dose range 10-100 Gy, then exposed to alpha particles from Am^{241} source. Figure (1) shows the variation of track density against the pre gamma dose. From the figure, it is obvious that the track density increases up to a maximum value around the 20 Gy irradiated sample, then almost decreases with increasing the gamma dose up to 100 Gy.

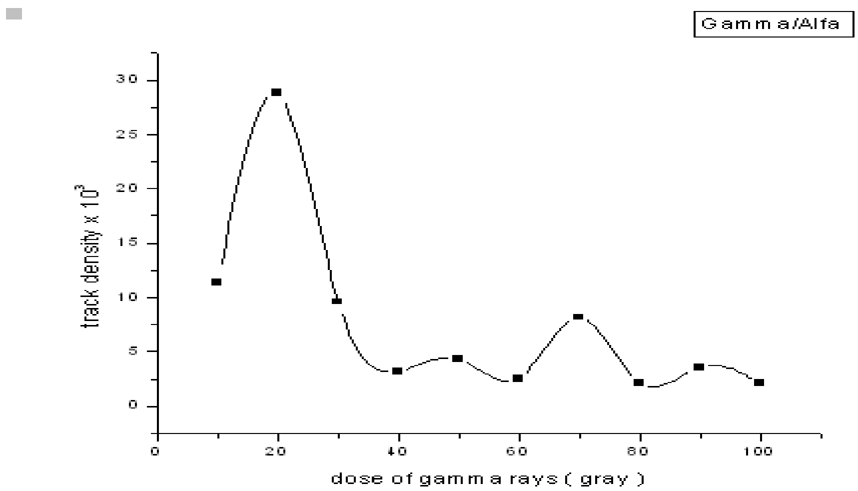

Figure (1) Variation of track density with the pre gamma dose

Post gamma irradiation:

Another group of samples were irradiated with alpha particles, and then exposed to gamma irradiation with the same doses.

Figure (2) shows the variation of track density with the post gamma dose. From the figure, it is clear that the track density increases up to a maximum value around the 40 Gy irradiated sample then decreases around the 50 Gy irradiated sample due to track overlapping. Above 50 and up to 100 Gy it increased again.

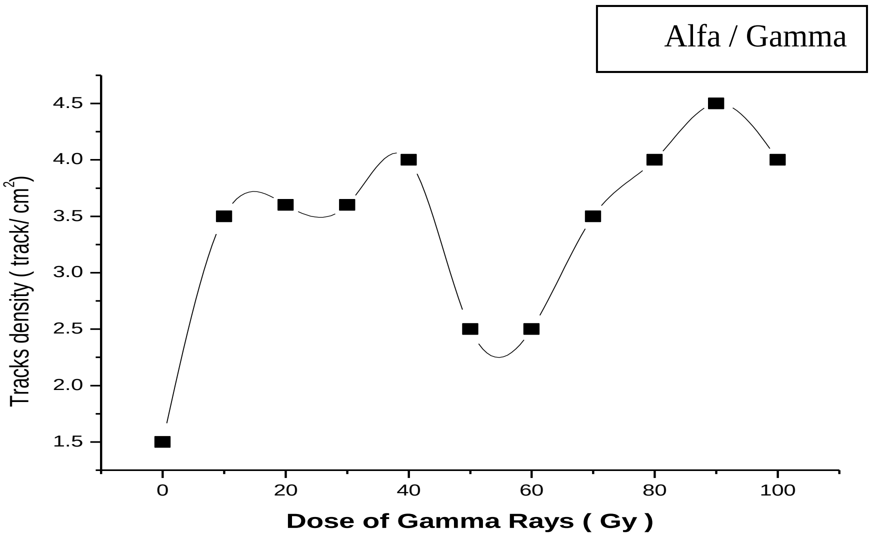

Figure (2) Variation of track density with the post gamma dose

The radon gas concentration in fertilizer samples was obtained by using the sealed-cup technique as shown in Figure 3. The irradiated CR-39 detectors were etched in 6.25 N NaOH at temperature of 70 ^{0}C for 6 h. Then the tracks density were recorded using an optical microscope with magnification 400x.The tracks density ρ in the samples were calculated according to the following relation (Amalds *et.al.*, 1989].

$$\mathbf{C} = \frac{\rho}{Kt} \tag{1}$$

Where ρ, *t* and *K* are the track density, the exposure time in hours and calibration factor $K = 0.16$ α-tracks $cm^{-2}.d^{-1}$ per $Bq.m^{-3}$ of radon. The track density can be calculated from the following relation

$$\text{The track Density } (\rho) \text{ (track } cm^{-2} \text{)} = \frac{\textit{Average of total tracks}}{\textit{Area of field of view}} \tag{2}$$

$$\rho = \frac{N - N_B}{S\,M}$$

where N, N_B, M and S are the number of tracks, background, number of fields and area of view, respectively. The radon gas concentration in the fertilizer samples were obtained by the comparison between track densities registered on the detectors before and after irritation. The sample was placed in sealed cup at bottom of cylindrical sealed can of 6 cm diameter and 14 cm height. The mouth of cylindrical can was sealed with cover and fitted with enhanced CR-39 plastic track detector (1cm x 1cm) at top inner surface. The detector records the tracks of α – particles emitted by radon gas produced through the α – decay of radium contents of the samples. The detectors were exposed for a period of about 90 days. After exposure, the detectors were retrieved and etched for six hours in 6.25 N Na OH solution maintained at temperature of 70 ^{0}C in a constant water bath to reveal the tracks.

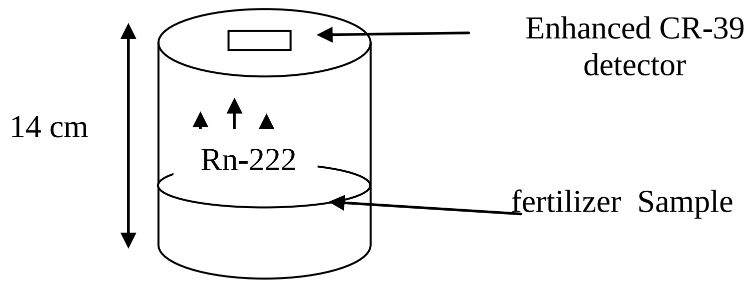

Figure (3): A schematic diagram of the sealed –cup technique in fertilizer sample.

Calculation of radon exhalation rate in fertilizer samples:

The radon exhalation rate of any sample is defined as the flux of radon released from the surface of material. The exhalation rate (E_A) in unit Bq m^{-2} h^{-1} can be calculated by (Mahur, 2010)

$$E_A = \frac{C\,V\,\lambda}{A\,[\,T + \lambda^{-1}(\,e^{-\lambda T} - 1)\,]} \qquad (3)$$

Where

C: is the integrated Radon concentration (Bq.m^{-3}).

V: is the effective volume of air in cup (m^3) =0.0001997 m^3

λ :is the decay constant for ^{222}Rn (h^{-1}) = 0.1812 day^{-1} =0.00755 h^{-1}

A: is the surface area of the sample (m^2) = 3^2×3.14=28.26 cm^2=0.002826 m^2

T: is the exposure time = 90 day and $T_{eff.}$ = 84.45 day $[\,T + \lambda^{-1}(\,e^{-\lambda T} - 1)\,]$

The annual effective dose (H_E) is calculated using the following expression: (UNSCEAR, 2000)

$$H_E\ (\,mSv\ y^{-1}\,) = C_{Rn} \times F \times H \times T \times D \qquad (4)$$

Where F is the equilibrium factor and it is equal to 0.4, H is the occupancy factor which is equal to 0.8, T is the time in hours in a year (8760 h y^{-1}) and (D) is the dose conversion factor which is equal to [9×10^{-6} (m Sv)/(Bq.h.m^{-3})] .

The lung cancer cases per year per million person (CPPP), was obtained using the relation (CPPP) = $H_E \times (18\times10^{-6}$ mSv^{-1}. y) (5)

RESULTS AND DISCUSSION

The interpretation of these results can be that the gamma irradiation of CR-39 in the dose range up to 20Gy causes chain scission. The free radicals produced from scission are chemically active and can cause crosslinking on gamma irradiation at the dose range 20-100 Gy. It is well known that track registration properties of the latent tracks are affected by exposing such detectors to gamma radiation (Jain, et.al, 2014 and Emad, *et.al*, 2015). The present findings clearly indicate the increasing relation between the bulk etching rate and gamma dose agree with that previously reported by *(*|Abu-Jarad *et al.*, 1997). The behavior of the removal thickness percentage with etching time is in agreement with that previously reported by (Malik *et al.*, 2002) The increasing relation between the removal thickness percentage and etchant concentration may be related to the attack by hydroxide ion results in the hydrolysis of the carbonate ester bonds and the release of polyallyalcohol from the polymer network (Brydson, 1975). The damage that appears in Figure(2) according to the degradation caused by the gamma rays interaction with the samples in addition to the etchant solution attack with the polymeric material of samples (Kuleznev and Shershnev, 1990). When ionizing particle passes through a plate of plastic, it produces a latent track as a result of damage caused by the energy deposition of the particle. This track can be made optically visible by means of chemical etching. Table (1), presents the Radon gas concentration, effective dose and exhalation rates for different kinds of fertilizer samples. The table illustrates that the highest average radon gas concentration in fertilizer samples was found in raw material equal to 269.620 Bq/m^3 then triple super phosphate (T.S.P), which was (186.8 Bq/m^3), while the lowest average radon gas concentration in fertilizer samples was found in single super phosphate (S.S.P), which was (79.6 Bq/m^3). Also the effective dose and radon exhalation rate for raw material and T.S.P. are higher than that for S.S.P. and waste product The present results show that all values in all fertilizer samples are below the allowed limit from the International Commission of Radiation Protection (ICRP) agency.

Table 1: The Radon gas concentration, Effective dose and Exhalation rates for different kinds of fertilizer samples

sample	Track density/Cm^2	Rn222 Conc.Bqm^{-3}	Effective dose mSv/y	Exhalation rates Bq m^{-2} h^{-1}
T.S.P		k.t=0.168*90 =15.12	D=**CR*0.4*.8*760*9*10$^{-6=25228.8}$**	
S1	2940	194.44	4.86	0.103
S2	2760	182	4.55	0.09
S3	2820	186.5	4.66	0.099
Average		186.8	4.712	0.097
S.S.P .powder				
S1	1140	75	1.89	0. 040
S2	1260	83.3	2.08	0.044
S3	1220	80.5	2.01	0.042
Average		79.6	2.00	0.042
S.S.P. Granules				

Table 1 (contin.)

sample	Track density/Cm^2	Rn222 Conc.Bqm^{-3}	Effective dose mSv/y	Exhalation rates Bq m^{-2} h^{-1}"
S1	760	62.16	1.568	0.026
S2	720	47.61	1.19	0.025
S3	740	48.94	740	0.026
Average		52.90	1.33	0.025
Raw material				
S1	4000	264.5	6.61	0.144
S2	4110	271.82	6.79	0.144
S3	4120	272.8	6.82	0.144
Average	4076.66	269.620	6.74	0.144
Waste product				
S1	1380	92	2.32	0.048
S2	1320	87.30	2.18	0.046
S3	1300	85.97	2.14	0.045
Average	1333.33	88.12	2.22	0.046
phosphogypsum				
S1	1060	70.1	1.75	0.037
S2	1000	66.3	1.65	0.035
S3	980	64.4	1.60	`0.034
Average	1013.33	67.01	1.69	0.035

CONCLUSION

The radon concentration , effective dose and exhalation rate for different fertilizer samples products from Abo-Zabal , and Kafer Alzyat factories were found that the maximum average values of raw material for radon concentration, effective dose ,exhalation rate were 269.62 Bq/m^3, 6.74 mSv/y, 1.44 Bq m^{-2} h^{-1} respectively and T.S.P was found that 186.8 Bq/m3, 4.712 mSv/y, 0.079 Bq m^{-2} h^{-1} respectively and lower average values for single super phosphate (S.S.P.) powder were 79.6 Bq/m^3, 2.0 mSv/y, 0.042 Bq m^{-2} h^{-1} respectively, S.S.P granules 52.90 Bq/m^3 , 1.33 mSv/y, 0.025 Bq

m^{-2} h^{-1} respectively and wast product average values were 88.12 Bq/m^3, 2.22 mSv/y, 0.046 Bq m^{-2} h^{-1} , phosphogypsium average values were 67.01 Bq/m^3, 1.69 mSv/y, 0.035 Bq m^{-2} h^{-1} respectively. All values are lower than the recommendation of International Commission on Radiological Protection range, (ICRP,1993)

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قياس تركيز غاز الرادون فى عينات السماد باستخدام كاشف الأثر النووى المعدل بالتشعيع

[٢]

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المستخلص

فى هذا البحث تم قياس تركيزات غاز الرادون فى عينات المواد الخام الفوسفاتية وعينات من سوبر الفوسفات الثلاثى باستخدام كاشف الأثر النووى وذلك بعد تحسين خواصه باستخدام أشعة جاما عند جرعة 20 Gy قبل تعريض هذه العينا ت لأشعة الفا.

وقد أوضحت الدراسة أن أعلى متوسط لتركيز غاز الرادون كان فى عينات مواد الخام الفوسفاتية حيث كانت متوسط تركيز غاز الرادون $269.62\ Bq/m^3$ بينما كان متوسط تركيز غاز الرادون فى عينات سوبر فوسفات الثلاثى $186.8\ Bq/m^3$ بينما كانت الجرعة الاشعاعية لخام الفوسفات 7.48 mSv/y وكان متوسط الجرعة الاشعاعية لسوبر الثلاثى 4.71 mSv/y بينما كان اقل قيمة لمتوسط غاز الرادون لسوبر الفوسفات الاحادى فكانت $79.2\ Bq/m^3$ ومتوسط الجرعة الاشعاعية لسوبر الفوسفات الاحادى كانت 2.008 mSv/y وكانت انبعاثات غاز الرادون لعينات الفوسفات محل الدراسة تتراوح من $0.025\ Bq\ m^{-2}h^{-1} – 0.144\ Bq\ m^{-2}\ h^{-1}$ وكانت النتائج للعينات محل الدراسة فى المدى المسموح به للوكالة الدولية للوقاية من الاشعاع